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U.S. PTO

Case Docket No. 108910-00011
Date: July 7, 2000

COMMISSIONER FOR PATENTS
Washington, D.C. 20231

Sir:

Transmitted herewith for filing under 37 C.F.R. §1.53(b) is the patent application of:

Inventor: Julio ABUSLEME and Claudia MANZONI

For: SYNTHESIS OF PERHALOGENATED THERMOPLASTIC (CO)POLYMERS OF CHLOROTRIFLUOROETHYLENE

- Specification (19 pages)
 Declaration and Power of Attorney attached.
 Return Receipt Postcard
 An assignment of the invention
 A certified copy of Italian application No. MI 99 A 001516 with English language translation
 Preliminary Amendment
 A filing fee, calculated as shown below:

(Col. 1)	(Col. 2)
FOR:	No. Filed
BASIC FEE	
TOTAL CLAIMS	16 - 20 = * 0
INDEP CLAIMS	1 - 3 = * 0
<u>0</u> MULTIPLE DEPENDENT CLAIM PRESENTED	

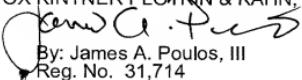
* If the difference in Col. 1 is less than zero, enter "0" in Col. 2

Small Entity	Other Than A Small Entity
RATE	RATE
\$345	\$690
or	or
× 9 =	× 18 =
or	or
× 39 =	× 78 =
or	or
+130 =	+260 =
TOTAL	
	\$690

- A check in the amount of \$730.00 to cover the filing fee (\$690.00) and Assignment recordation (\$40.00) is enclosed.
- In the event that the attached check is found to be insufficient, the Commissioner is hereby authorized to charge payment for any additional filing fees required under 37 CFR 1.16 associated with this communication or credit any over-payment to Deposit Account No. 01-2300.

Respectfully submitted,

AREN'T FOX KINTNER PLOTKIN & KAHN, PLLC


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J-564 U.S. PTO
09/61287
07/07/00

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of:

J. ABUSLEME et al.

Serial Number: not yet assigned

Group Art Unit: not yet assigned

Filed: July 7, 2000

Examiner: not yet assigned

For: SYNTHESIS OF PERHALOGENATED THERMOPLASTIC (CO)POLYMERS
OF CHLOROTRIFLUOROETHYLENE

PRELIMINARY AMENDMENT

Commissioner for Patents
Washington, D.C. 20231

July 7, 2000

Sir:

Prior to calculation of the Filing Fee and examination of the above-identified U.S. patent application on the merits, please enter the following amendments.

IN THE CLAIMS:

Please amend the claims as indicated below.

Claim 4, line 1, change "claims 1-3" to --claim 1 ---.

Claim 6 line 1, change "claims 1-5" to --claim 1 --.

Claim 8, line 1, change "claims 1-7" to --claim 10 --.

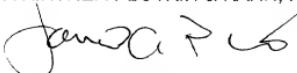
REMARKS

The above amendments to the claims have been made to correct multiple dependency of the claims and to put the application in better condition for examination.

In view of the amendments and remarks above, Applicants submit that this application is in condition for examination and respectfully request an early action on the merits. In the event any fees are required with respect to this paper, please charge our Deposit Account No. 01-2300.

Respectfully submitted,

AREN'T FOX KINTNER PLOTKIN & KAHN, PLLC



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SYNTHESIS OF THERMOPLASTIC PERHALOGENATED (CO)POLYMERS OF
CHLOROTRIFLUOROETHYLENE

ABSTRACT

A process for the synthesis of chlorotrifluoroethylene (PCTFE) (co)polymers, containing at least 80% by moles of CTFE, being the complement to 100 one or more fluorinated monomers, preferably the complement to 100 is formed of one or more perfluorinated monomers, characterized in that the reaction medium comprises (per)fluoropolyoxyalkylene microemulsions wherein the fluorinated surfactant is salified with sodium and/or potassium and an inorganic potassium and/or sodium initiator.

The present invention relates to a process for obtaining halogenated polymers having good thermal stability.

Specifically the present invention relates to a process for obtaining chlorotrifluoroethylene (CTFE) polymers having substantially no discoloration, combined with a good thermal stability.

The chlorotrifluoroethylene (PCTFE) polymers according to the process of the present invention contain at least 80% by moles of CTFE and the complement to 100 being one or more fluorinated monomers, preferably the complement to 100 is formed of one or more perfluorinated monomers.

The polymers obtained in the process of the invention are to be used for manufacturing pipes, sheets, films and other articles having the advantages indicated below.

More specifically it is known in the prior art that the CTFE homopolymer is a fluorinated resin having excellent chemical resistance with good properties of impermeability to gases and vapours, more specifically to oxygen, nitrogen and water vapour, which make this resin the most suitable material for the extrusion in films for the pharmaceutical packaging

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industry.

For the above mentioned applications, it is desirable to use a PCTFE having good thermal stability and substantially no discoloration, allowing a wide processing window, i.e. high temperatures and long residence times in extrusion such that the PCTFE does not undergo discoloration without substantial weight loss. This wide processing window allows to obtain articles having complex shapes which require long residence times and high temperatures. In particular, the availability of a PCTFE with a wide processing window could allow the processing of polymers of high molecular weight and therefore to manufacture articles with improved mechanical properties from those PCTFE's of low molecular weight.

It is therefore desirable to have available an efficient industrial polymerization process, i.e. with yields not lower than about 10 g/(l×h) (g of polymer/l of water.h), thermodynamically stable latexes and absence of polymer buildup in the polymerization reactor, which allows the PCTFE synthesis of different viscosity values, in particular high molecular weight PCTFE's, having good thermal stability combined with substantial no discoloration.

The Applicant has unexpectedly and surprisingly found an efficient process for obtaining the above PCTFE's.

It is therefore an object of the present invention a

process for the synthesis of chlorotrifluoroethylene (PCTFE) (co)polymers, containing at least 80% by moles of CTFE, being the complement to 100 one or more fluorinated monomers, preferably the complement to 100 is formed of one or more perfluorinated monomers, characterized in that the reaction medium comprises (per)fluoropolyoxyalkylene microemulsions wherein the fluorinated surfactant is salified with sodium and/or potassium and an inorganic potassium and/or sodium initiator.

The (per)fluoropolyoxyalkylene microemulsions according to the present invention are obtainable following the methods described in USP 4,864,006, in the name of the Applicant, herein incorporated by reference.

The fluorinated surfactants of the microemulsions according to the present invention are selected from the products of general formula



wherein R_f , is a C_5-C_{14} (per)fluoroalkyl chain, or a (per)fluoropolyoxyalkylene chain, X^+ is $-COO^-$ or $-SO_3^-$, M^+ is selected between Na^+ and K^+ , preferably K^+ .

According to the present invention, potassium inorganic initiators are preferred, potassium persulphate is still more preferred.

The process of the invention is carried out at a

temperature between 0°C and 150°C, preferably between 10°C and 70°C. The reaction pressure is generally in the range 3-80 bar, preferably 4-20 bar.

The amount of the radical usable initiator is the standard one for the copolymerization of fluorinated olefinic monomers, and is generally in the range 0.003%-10% by weight with respect to the total amount of (co)polymerized monomers.

The control of the molecular weight of the fluorinated polymers of the invention can be carried out in various ways.

Preferred are those by a suitable dosage of the radical initiator in polymerization and the selection of the synthesis temperature. To obtain the polymers of the invention having high molecular weight, i.e. having MFI lower than 15 g/10', a low synthesis temperature (10°C-50°C) combined with a low concentration of free radicals coming from the initiator is preferred. To obtain the invention polymers having low molecular weight, i.e. having a MFI higher than 5 g/10', a high synthesis temperature (60°C-80°C) combined with a high concentration of free radicals coming from the initiator, is preferred.

Another way to control the molecular weight is the use of chain transfer agents, these can be halogenated hydrocarbons, for example chloroform or HCFC 123 and ethane or methane. The transfer agent is fed to the reactor at the

beginning of the reaction, or in a continuous way or in discrete amounts during the polymerization. The used chain transfer agent amount can range within rather wide limits, depending on the reaction temperature and the molecular weight target. Generally, such amount ranges from 0.001 to 5% by weight, preferably from 0.05 to 1% by weight, with respect to the total amount of monomers fed to the reactor.

In the process according to the present invention the presence of liquid CTFE in the reaction medium, in order to obtain high productivity (R_p) in g/(1×h), is preferred.

A further object of the present invention are chlorotrifluoroethylene (PCTFE) (co)polymers, obtainable by the above described process, containing at least 80% by moles of CTFE, being the complement to 100 one or more fluorinated monomers, preferably the complement to 100 being one or more perfluorinated monomers.

The chlorotrifluoroethylene (PCTFE) (co)polymers of the invention are characterized in that the CTFE homopolymer, having a MFI of about 25 g/10', does not show discoloration, as defined below, and a global very reduced weight loss by thermogravimetric analysis.

The preferable (co)polymers of the invention are the thermoplastic ones.

The CTFE homopolymers with MFI of about 25 g/10', obtai-

ned according to the present invention, have a weight loss which is approximately the half of the weight loss of the CTFE homopolymers obtained in emulsion.

Besides the efficiency confirmation of the process object of the present invention is mainly given by the high productivity yield and by the absence of polymer buildup in autoclave.

This low weight loss of the polymers of the invention combined with absence of discoloration allows to obtain manufactured articles of complex shape and with substantial absence of defects, mainly bubbles. For example, the presence of bubbles in the extruded film makes it unusable, since the main PCTFE characteristic is the impermeability.

Among the fluorinated monomers, we can mention hexafluoropropylene, hexafluoroisobutylene, vinylidenefluoride, tetrafluoroethylene, fluorinated ethers such as perfluoroalkylvinylethers, for example perfluoroethylvinylether, perfluoromethylvinylether, preferably perfluoropropylvinyl-ether, or mixtures thereof, .

The viscosity of CTFE (co)polymers according to the present invention is defined by the Melt Flow Index measure (MFI) at 265°C and 10 Kg load according to the ASTM D 1238-88 method.

The discoloration is determined through the observation

of the strands obtained after 20, 40 and 60 minutes of residence time of the polymer in the MFI machine at 265°C. Discoloration means the coloration of the extruded polymer. The product not showing discoloration is colourless or white. Polymers showing discoloration are generally yellowish or brown. The skilled in the art is able to determine whether the polymer shows or not discoloration also by using methods measuring the colour index such as for example white index or yellow index.

The thermal stability is subsequently controlled, if there is substantially no discoloration as above defined, through thermo-gravimetric measures (TGA) carried out as follows:

- i) Dynamic TGA with a heating rate of 20°C/minute up to 300°C;
- ii) Isotherm TGA at 300°C for 60 minutes.

The global variations by weight (ΔW , % by weight) and the weight loss rate in the last 10 minutes of the isothermal treatment ($\Delta W/\Delta t$, % by weight/h) are determined (see Table 1).

The perfluoropropylvinylether content in the PCTFE copolymers has been determined by $^{19}\text{F-NMR}$.

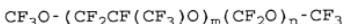
The second melting temperature (T_{2f}) and the crystallization temperature (T_{xx}) are determined by differential scanning calorimetry (DSC).

The following Examples are given for illustrative purposes and are not limitative of the present invention.

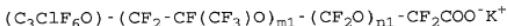
EXAMPLE 1

A 2 l AISI 316 autoclave, equipped with stirrer working at 400 rpm, has been evacuated and therein were introduced in sequence:

- 1300 g of demineralized H₂O;
- 14 g of a microemulsion formed of: 20% by weight of Gal-den® D02, having formula:



having m/n = 20 and average molecular weight of 450; 40% by weight of a surfactant having formula:



having m1/n1 = 82.7 and average molecular weight of 527; the remaining part being formed of H₂O.

In the autoclave 400 g of CTFE were then introduced between 1°C and 5°C. Then the autoclave was heated to the reaction temperature of 60°C and 3.0 g of potassium persulphate dissolved in 90 g of demineralized H₂O were then introduced.

After 240 minutes of reaction, when the working pressure has decreased of 50% with respect to its initial value, the autoclave is vented and discharged at room temperature.

The latex discharged from the autoclave, having a con-

centration of 250.0 g/l of water, was cryogenically coagulated, then the polymer was separated and dried at 175°C for about 16 hours.

After the latex was discharged and the autoclave washed with water, its internal walls did not show residues.

No discoloration of the strands obtained after 60 minutes of residence time in the MFI machine at 265°C, was noticed.

The test result of the thermal stability of the obtained polymer is shown in Table 1. As a further proof of the thermal stability no discoloration of the strands obtained after 80 minutes of residence time in the MFI machine at 280°C, was noticed.

EXAMPLE 2 comparative (comp)

A 2 l AISI 316 autoclave, equipped with stirrer working at 400 rpm, has been evacuated and therein were introduced in sequence:

- 1200 g of demineralized H₂O;
- a solution of 5.6 g of sodium perfluoroctanoate (PFONa) in 100 g of water, corresponding to the surfactant amount fed in Example 1.

In the autoclave 400 g of CTFE were then introduced between 1°C and 5°C. Then the autoclave was heated to the reaction temperature of 60°C and 3.0 g of potassium persulphate dissolved in 90 g of demineralized H₂O were then introduced.

After 705 minutes of reaction, when the working pressure has decreased of 50% with respect to its initial value, the autoclave is vented and discharged at room temperature.

The latex discharged from the autoclave, having a concentration of 239.0 g/l of water, was cryogenically coagulated, then the polymer was separated and dried at 175°C for about 16 hours.

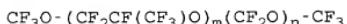
After having discharged the latex and washed with water the autoclave, its internal walls show some residues.

No discoloration of the strands obtained after 60 minutes of residence time in the MFI machine at 265°C, was noticed.

EXAMPLE 3 comparative (comp)

A 2 l AISI 316 autoclave, equipped with stirrer working at 400 rpm, has been evacuated and therein were introduced in sequence:

- 1300 g of demineralized H₂O;
- 18.3 g of a microemulsion formed of: 18.37% by weight of Galden® D02, having formula:



having m/n = 20 and average molecular weight of 450;

30.61% by weight of a surfactant having formula:



having m1/n1 = 82.7 and average molecular weight of 527, corresponding to the surfactant amount introduced in

Example 1; the remaining part being formed of H₂O.

In the autoclave 400 g of CTFE were then introduced between 1°C and 5°C. Then the autoclave was heated to the reaction temperature of 60°C and 2.5 g of ammonium persulphate, equivalent by moles to the potassium persulphate of Example 1, dissolved in 90 g of demineralized H₂O were then introduced.

After 195 minutes of reaction, when the working pressure has decreased of 50% with respect to its initial value, the autoclave is vented and discharged at room temperature.

The latex discharged from the autoclave, having a concentration of 259.0 g/l of water, was cryogenically coagulated, then the polymer was separated and dried at 175°C for about 16 hours.

After the latex was discharged, part of this is present in the form of coagulum on the bottom of the autoclave.

Discoloration of the obtained strands was noticed already after 20 minutes of residence time in the MFI machine at 265°C.

EXAMPLE 4 comparative (comp)

The comparative Example 3 was repeated, replacing the ammonium persulphate with 3.0 g of potassium persulphate.

After 245 minutes of reaction, when the working pressure has decreased of 50% with respect to its initial value, the autoclave is vented and discharged at room temperature.

The latex discharged from the autoclave, having a concentration of 260 g/l of water, was cryogenically coagulated, then the polymer was separated and dried at 175°C for about 16 hours.

After the latex was discharged and the autoclave washed with water, its internal walls did not show residues.

Discoloration of the obtained strands was noticed already after 20 minutes of residence time, in the MFI machine at 265°C.

EXAMPLE 5 comparative (comp)

Example 1 was repeated, replacing the potassium persulphate with an equivalent amount by moles of ammonium persulphate: 2.5 g.

After 272 minutes of reaction, when the working pressure has decreased of 50% with respect to its initial value, the autoclave is vented and discharged at room temperature.

The latex discharged from the autoclave, having a concentration of 275 g/l water, was cryogenically coagulated, then the polymer was separated and dried at 175°C for about 16 hours.

After the latex was discharged and the autoclave washed with water, its internal walls did not show residues.

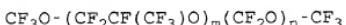
Discoloration of the obtained strands was noticed already after 20 minutes of residence time in the MFI machine at

265°C.

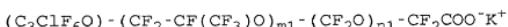
EXAMPLE 6

A 2 l AISI 316 autoclave, equipped with stirrer working at 400 rpm, has been evacuated and therein were introduced in sequence:

- 1200 g of demineralized H₂O;
- 14 g of a microemulsion formed of: 20% by weight of Gal-den® D02, having formula:



having m/n = 20 and average molecular weight of 450; 40% by weight of a surfactant having formula:



having m1/n1 = 82.7 and average molecular weight of 527; the remaining part being formed of H₂O

- 3.0 g of potassium persulphate dissolved in 200 g of demineralized H₂O.

In the autoclave 33 g of perfluoropropylvinylether (FPVE) and 400 g of CTFE were then introduced between 1°C and 5°C.

Then the autoclave was heated to reaction temperature of 50°C.

After 500 minutes of reaction, when the working pressure has decreased of 50% with respect to its initial value, the autoclave is vented and discharged at room temperature.

The latex discharged from the autoclave, having a con-

centration of 268 g/l of water, was cryogenically coagulated, then the polymer was separated and dried at 175°C for about 16 hours.

The perfluoropropylvinylether content in the obtained copolymer is 1.5% by moles.

After the latex was discharged and the autoclave was washed with water, its internal walls did not show residues.

No discoloration of the obtained strands was noticed after 60 minutes of residence time in the MFI machine at 265°C.

EXAMPLE 7 comparative

A 2 l AISI 316 autoclave, equipped with stirrer working at 400 rpm, has been evacuated and therein were introduced in sequence:

- 1200 g of demineralized H₂O;
- a solution of 5.6 g of sodium perfluoroctanoate (PFONa) in 100 g of water, corresponding to the surfactant amount introduced in Example 1.
- 3.0 g of potassium persulphate dissolved in 200 g of demineralized H₂O.

In the autoclave 33 g of perfluoropropylvinylether (FPVE) and 400 g of CTFE were then introduced between 1°C and 5°C. Then the autoclave was heated to the reaction temperature of 50°C.

After 1890 minutes of reaction, when the working pressure has decreased of 50% with respect to its initial value, the autoclave is vented and discharged at room temperature.

The latex discharged from the autoclave, having a concentration of 239 g/l in water, was cryogenically coagulated, then the polymer was separated and dried at 175°C for about 16 hours.

After the latex was discharged and the autoclave washed with water, its internal walls are completely covered by a layer of the obtained polymer.

Discoloration of the obtained strands was noticed after 60 minutes of residence time in the MFI machine at 265°C.

TABLE 1

EXAMPLE	1	2 (comp)	6	7 (comp)
T _{2f} (°C)	212.2	211.4	197.1	--
T _{xx} (°C)	181.7	175.5	162.0	--
R _p (g/(l×h))	62.5	20.3	32.2	7.6
MFI(g/10')	25.0	24.6	9.8	4.9
ΔW (% w)	0.34	0.81	--	--
ΔW/Δt (% w/h)	0.126	0.312	--	--
DISCOLORATION	NO	NO	NO	YES

CLAIMS

1. A process for the synthesis of chlorotrifluoroethylene (PCTFE) (co)polymers, containing at least 80% by moles of CTFE, the complement to 100 being one or more fluorinated monomers, preferably the complement to 100 is formed of one or more perfluorinated monomers, characterized in that the reaction medium comprises (per)fluoropolyoxyalkylene microemulsions wherein the fluorinated surfactant is salified with sodium and/or potassium and an inorganic potassium and/or sodium initiator.
2. A process according to claim 1, wherein the fluorinated surfactant is selected from the products of general formula



wherein R_f is a C_5-C_{14} (per)fluoroalkyl chain, or a (per)fluoropolyoxyalkylene chain, X^- is $-COO^-$ or $-SO_3^-$, M^+ is selected between Na^+ and K^+ .

3. A process according to claim 2, wherein M^+ is preferably K^+ .
4. A process according to claims 1-3, wherein potassium inorganic initiators are preferred.
5. A process according to claim 4, wherein the potassium inorganic initiator is potassium persulphate.
6. A process according to claims 1-5, wherein the tempera-

- ture is in the range 0°C-150°C and the pressure is in the range 3-80 bar.
7. A process according to claim 6, wherein the temperature ranges between 10°C and 70°C and the pressure between 4 and 20 bar.
 8. A process according to claims 1-7, wherein the presence of liquid CTFE in the reaction medium is preferred.

2025 RELEASE UNDER E.O. 14176

Declaration For U.S. Patent Application

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below my name.

I believe I am the original, first sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled
(Insert Title) **"SYNTHESIS OF THERMOPLASTIC PERHALOGENATED (CO)POLYMERS
OF CHLOROTRIFLUOROETHYLENE"**

the specification of which is attached hereto unless the following box is checked:

was filed on _____ as PCT International Application
Number _____ and was amended on _____
and/or was filed on _____ as United States Application
Number _____ and was amended on _____.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claim(s), as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 C.F.R. §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below any foreign application for patent or inventor's certificate or PCT International Application having a filing date before that of the application(s) for which priority is claimed:

MT99 A 001516	ITALY	09 JULY 1999	Priority Claimed
(List prior foreign applications. See note A on back of this page)	(Country)	(Day/Month/Year Filed)	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
(Number)	(Country)	(Day/Month/Year Filed)	<input type="checkbox"/> Yes <input type="checkbox"/> No
(Number)	(Country)	(Day/Month/Year Filed)	<input type="checkbox"/> Yes <input type="checkbox"/> No

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below.

(Application Number) _____ (Filing Date) _____

(Application Number) _____ (Filing Date) _____

(See Note B on back of this page)

See attached list for additional prior foreign or provisional applications.

I hereby claim the benefit under 35 U.S.C. §120 of any United States application(s) or §365(c) of any PCT International application(s) designating the United States of America listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior application(s) (U.S. or PCT) in the manner provided by the first paragraph of 35. U.S.C. §112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 C.F.R. §1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

(List prior U.S. Applications or PCT International applications designating the U.S.)	(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)
	(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)

And I hereby appoint as principal attorneys: David T. Nikaido, Reg. No. 22,663; Charles M. Marmelstein, Reg. No. 25,895; George E. Oran, Jr., Reg. No. 27,931; Robert B. Murray, Reg. No. 22,980; E. Marcie Emas, Reg. No. 32,131; Douglas H. Goldhush, Reg. No. 33,125; Monica Chin Kitts, Reg. No. 36,105; Richard J. Herman, Reg. No. 39,107; King L. Wong, Reg. No. 37,500; James A. Poulos, III, Reg. No. 31,714; Patrick D. Muir, Reg. No. 37,403; Sharon N. Kleiner, Reg. No. 36,335; Murat Ozgu, Reg. No. 44,275; Bradley D. Goldizen, Reg. No. 43,637, and N. Alexander Nolte, Reg. No. 45,689.

Please direct all communications to the following address: ARENT FOX KINTNER PLOTKIN & KAHN, PLLC
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Telephone No. (202) 857-6000; Facsimile No. (202) 857-6395

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

(See Note C on back of this page)

Full name of sole or first inventor	Julio A. ABUSLEME
Inventor's signature	
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Citizenship	Italian
Post Office Address	Via Bergamo 5 – 21047 SARONNO, Varese, Italy
Date	June 22, 2000

Full name of second joint inventor, if any	Claudia MANZONI	
Inventor's signature	<u>Claudia Manzoni</u>	June 22, 2000
Residence	BOLOGNA, Italy	Date
Citizenship	Italian	
Post Office Address	Via del Lino 3 - 40100 BOLOGNA, Italy	
Full name of third joint inventor, if any		
Inventor's signature		
Residence		
Citizenship		
Post Office Address		
Full name of fourth joint inventor, if any		
Inventor's signature		
Residence		
Citizenship		
Post Office Address		
Full name of fifth joint inventor, if any		
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Post Office Address		
Full name of seventh joint inventor, if any		
Inventor's signature		
Residence		
Citizenship		
Post Office Address		
Full name of eighth joint inventor, if any		
Inventor's signature		
Residence		
Citizenship		
Post Office Address		
Full name of ninth joint inventor, if any		
Inventor's signature		
Residence		
Citizenship		
Post Office Address		